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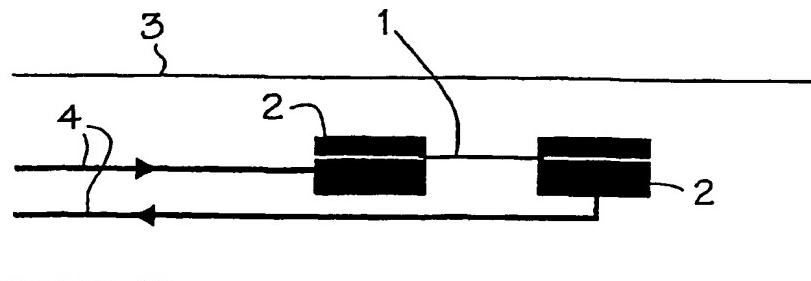
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(54) Title: PROCESS FOR PREPARING CARBON NANOTUBES

Ar + H₂ +
Hydrocarbon
→



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(57) Abstract: Carbon nanotubes are formed on carbon paper by first depositing a metal catalyst on the carbon paper, and passing a feedstock gas containing a source of carbon over the substrate while applying an electrical current thereto to heat the substrate sufficiently to generate a reaction between the catalyst and the feedstock gas. Alternatively, inert gas under pressure is passed through a tubular metal cathode while passing an electric current through the cathode to produce a plasma of fine catalyst particles which are deposited on a porous carbon substrate, and a feedstock gas containing a source of carbon is passed over the substrate to cause a reaction between the catalyst and the carbon source resulting in the formation of carbon nanotubes.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER
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C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 1999, no. 10, 31 August 1999 (1999-08-31) & JP 11 139815 A (CANON INC), 25 May 1999 (1999-05-25) abstract ---	1
A, P	EP 1 059 266 A (ILJIN NANOTECH CO LTD ;LEE CHEOL JIN (KR)) 13 December 2000 (2000-12-13) claim 1 --- -/-	1

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Patent family members are listed in annex.

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>QIN L C ET AL: "GROWING CARBON NANOTUBES BY MICROWAVE PLASMA-ENHANCED CHEMICAL VAPOR DEPOSITION" APPLIED PHYSICS LETTERS, AMERICAN INSTITUTE OF PHYSICS, NEW YORK, US, vol. 72, no. 26, 29 June 1998 (1998-06-29), pages 3437-3439, XP000771159 ISSN: 0003-6951 page 3437 -page 3438</p> <p>-----</p>	1

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Information on patent family members

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PCT/CA 01/00658

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
JP 11139815	A 25-05-1999	NONE		
EP 1059266	A 13-12-2000	CN 1277145 A		20-12-2000
		EP 1059266 A2		13-12-2000
		JP 2001020071 A		23-01-2001

Electrochemistry of Carbon Nanotubes and Their Potential Application in Supercapacitors

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ABSTRACT

Carbon nanotubes are grown on graphite sheet, carbon cloth and other materials by chemical vapor deposition (CVD) method. The electrochemical behavior of carbon nanotubes has been investigated by cyclic voltammetry in 1.0M H_2SO_4 aqueous solutions. High "effective capacitance" and rectangular-shaped cyclic voltammograms at high scan rate (100 mV/s) have been observed, which makes these carbon nanotubes of great interest as electrodes in supercapacitors. The material is very stable on cycling and no significant difference has been seen after continuous cycles over 30.

INTRODUCTION

Carbon nanotubes are very interesting materials in the sense of their structure and their size in the nanometer range, which leads to that they have highly accessible surface area, low resistivity, and high stability [1-3]. These features are ideally desired for carbon nanotubes to be used as electrodes in supercapacitors [4].

EXPERIMENTAL

Carbon nanotubes were grown directly on several substrates: graphite sheet, carbon cloth and graphite sheet covered with activated carbon by chemical vapor deposition (CVD) method using acetylene (C_2H_2) and ammonia (NH_3) [5-7]. Ni particles were deposited on the substrates and used as catalyst. Before the measurement of cyclic voltammogram, samples were immersed into 15% wt. HNO_3 aqueous solution for 30 minutes in order to remove the metallic particle (catalyst) and increase the wettability of the surface of the carbon nanotubes in aqueous solution. PC4 Potentiostat/Galvanostat (Gamry Instruments Inc. Warminster, PA 18974) was employed for the cyclic voltammetric (CV) measurements in 1.0M H_2SO_4 aqueous solutions. A platinum wire served as the counter electrode, and a saturated calomel electrode (SCE) was used as reference electrode.

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RESULTS and DISCUSSIONS

Carbon nanotubes were grown on three kinds of substrates: activated carbon-coated graphite foil, bare graphite foil, and carbon cloth. The SEM images of the carbon nanotubes are shown in Fig.1, Fig.2 and Fig.3 respectively. Nanotube bundles can be found in Fig.1 and Fig.3.



Figure 1. SEM images of carbon nanotubes grown directly on activated carbon-coated graphite foil. (a) low magnification to show the uniformity in large area and length; (b) high magnification to show the diameter.

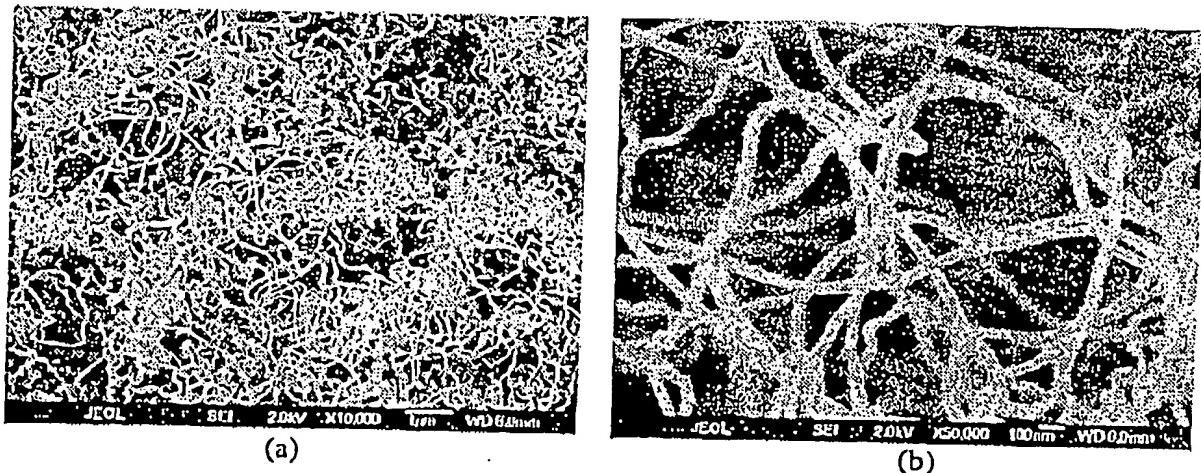


Figure 2. SEM images of carbon nanotubes grown directly on the graphite foil. (a) low magnification to show the uniformity in large area and length; (b) high magnification to show the diameter.

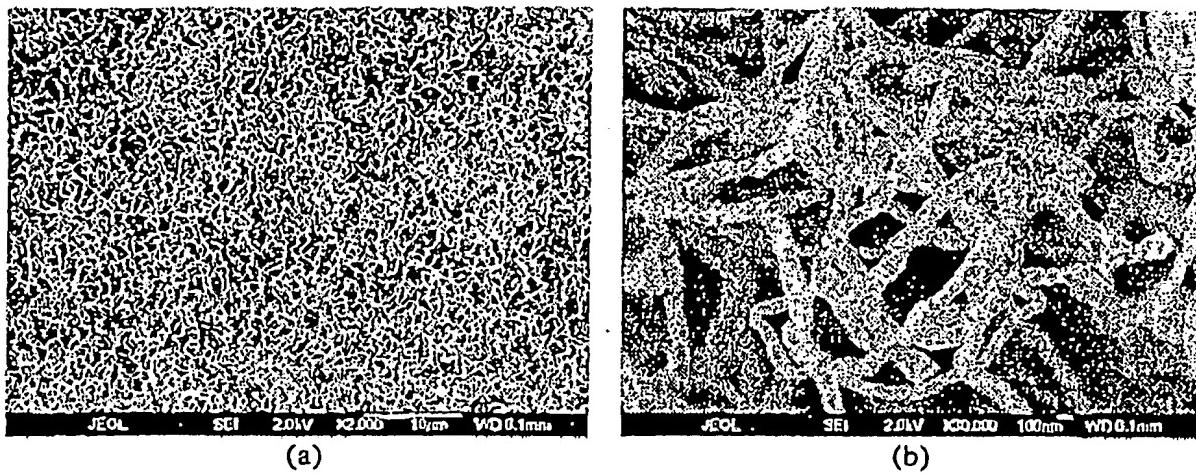


Figure 3. SEM images of carbon nanotubes grown directly on carbon cloth substrate. (a) low magnification to show the uniformity in large area and length; (b) high magnification to show the diameter.

The electrochemical properties of these nanotube electrodes were studied by cyclic voltammetry in 1.0M H_2SO_4 aqueous solution. The typical cyclic voltammograms (CVs) are shown in Fig.4, Fig.5, Fig.6 and Fig.7. These CVs are featureless voltammograms and no Faradic peaks can be observed between 0 to 0.9 V (vs. SCE). This result has also been observed for the single-walled carbon nanotubes [4].

Rectangular-shaped cyclic voltammograms over a wide range of scan rates is the ultimate goal in electrochemical double-layer capacitors. This behavior is very important for practical applications. In Fig.4, Fig.5, Fig.6 and Fig.7, These carbon nanotube electrodes can retain the rectangular shape of CVs up to a high scan rate (100mV/s). This means the charge and discharge processes are very fast at the interface between the nanotube electrode and electrolyte

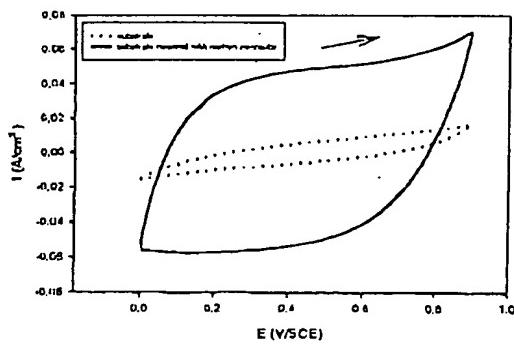


Figure 4. Cyclic voltammogram of sample #1 in 1.0M H_2SO_4 aqueous solution. Sample #1: carbon nanotubes were grown directly on the graphite sheet substrate covered with activated carbon. Scan rate: 100mV/s.

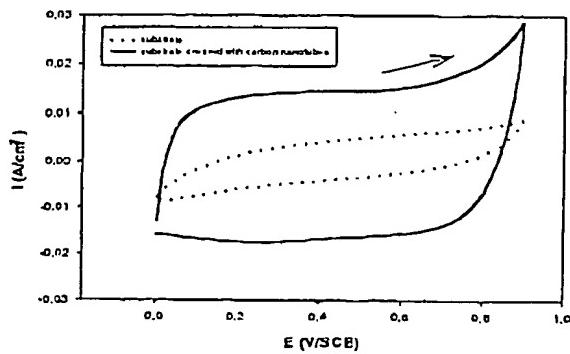


Figure 5. Cyclic voltammogram of sample #1 in 1.0M H_2SO_4 aqueous solution. Sample #1: carbon nanotubes were grown directly on the graphite sheet substrate covered with activated carbon. Scan rate: 25mV/s.

solution. The featureless CVs and high speeds of charge and discharge suggest a possible application of this kind of multi-walled carbon nanotubes to supercapacitor. The material is stable on cycling and no significant difference can be seen after continuous cycles over 30 cycles.

The results in Fig.4, Fig.5, Fig.6 and Fig.7 show that the electrochemical capacitance of the electrode increases obviously when carbon nanotubes are grown on the substrates. The mass of carbon nanotubes grown on the substrate is obtained from the difference of total weight of the electrode before and after the growth of carbon nanotubes. The capacitance is calculated from the CV curves, with $C = \int i dt / \Delta V$, where i is the current. The increases of the effective capacitance per unit weight of carbon nanotubes can be calculated, for example, at the scan rate of 25mV/s, are 98.6 F/g and 140 F/g for sample #1 and sample #2, respectively.

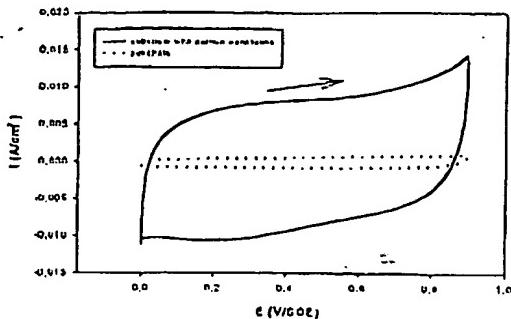


Figure 6. Cyclic voltammogram of sample #2 in 1.0M H_2SO_4 aqueous solution. Sample #2: carbon nanotubes were grown directly on the graphite sheet. Scan rate: 100mV/s

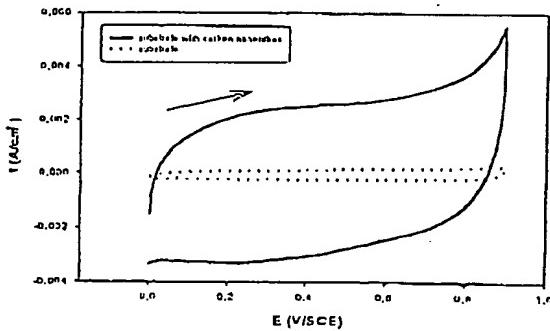


Figure 7. Cyclic voltammogram of sample #2 in 1.0M H_2SO_4 aqueous solution. Sample #2: carbon nanotubes were grown directly on the graphite sheet. Scan rate: 25mV/s

CONCLUSIONS

Carbon nanotubes are excellent electrodes for supercapacitors.

ACKNOWLEDGEMENTS

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